

FORM PTO-1390
(REV. 11-2000)

U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE

ATTORNEY'S DOCKET NUMBER

TRANSMITTAL LETTER TO THE UNITED STATES
DESIGNATED/ELECTED OFFICE (DO/EO/US)
CONCERNING A FILING UNDER 35 U.S.C. 371

270/170

U.S. APPLICATION NO. (If known, see 37 CFR 1.5)

10/031135

INTERNATIONAL APPLICATION NO.

INTERNATIONAL FILING DATE

PRIORITY DATE CLAIMED

PCT/EP00/06423

July 6, 2000

July 14, 1999

TITLE OF INVENTION

CURABLE COMPOSITION

APPLICANT(S) FOR DO/EO/US

Christoph RICKERT; Martin ROTH; Francois TURPIN; Jacques FRANCOIS; Klaus PETSCHER

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. ☒ This is a **FIRST** submission of items concerning a filing under 35 U.S.C. 371.
2. ☐ This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. 371.
3. ☐ This is an express request to begin national examination procedures (35 U.S.C. 371(f)). The submission must include items (5), (6), (9) and (21) indicated below.
4. ☒ The US has been elected by the expiration of 19 months from the priority date (Article 31).
 - ☒ A copy of the International Application as filed (35 U.S.C. 371(c)(2))
 - a. ☐ is attached hereto (required only if not communicated by the International Bureau)
 - b. ☒ has been communicated by the International Bureau
 - c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US).
 - 6. ☐ An English language translation of the International Application as filed (35 U.S.C. 371(c)(2)).
 - a. ☐ is attached hereto.
 - b. ☐ has been previously submitted under 35 U.S.C. 154(d)(4)
 - ☒ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3))
 - a. ☐ are attached hereto (required only if not communicated by the International Bureau).
 - b. ☐ have been communicated by the International Bureau.
 - c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.
 - d. ☒ have not been made and will not be made.
8. ☐ An English language translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).
9. ☐ An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)).
10. ☐ An English language translation of the annexes of the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).

Items 11 to 20 below concern document(s) or information included:

11. ☐ An Information Disclosure Statement under 37 CFR 1.97 and 1.98.
12. ☐ An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
13. ☒ A **FIRST** preliminary amendment.
14. ☐ A **SECOND** or **SUBSEQUENT** preliminary amendment.
15. ☐ A substitute specification.
16. ☐ A change of power of attorney and/or address letter.
17. ☐ A computer-readable form of the sequence listing in accordance with PCT Rule 13ter.2 and 35 U.S.C. 1.821 – 1.825.
18. ☒ A second copy of the published international application under 35 U.S.C. 154(d)(4).
19. ☐ A second copy of the English language translation of the international application under 35 U.S.C. 154(d)(4).
20. ☒ Other items or information: **PCT/IB/308**

U.S. APPLICATION NO. (if known, see 37 CFR 1.5) <div style="font-size: 24pt; font-weight: bold; margin-top: 5px;">10/031135</div>		INTERNATIONAL APPLICATION NO. PCT/EP00/06423		ATTORNEY'S DOCKET NUMBER 270/170	
21. <input checked="" type="checkbox"/> The following fees are submitted: BASIC NATIONAL FEE (37 CFR 1.492(a)(1) – (5)): Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO and International Search Report not prepared by the EPO or JPO \$1040.00 International preliminary examination fee (37 CFR 1.482) not paid to USPTO but International Search Report prepared by the EPO or JPO \$890.00 International preliminary examination fee (37 CFR 1.482) not paid to USPTO but international search fee (37 CFR 1.445(a)(2)) paid to USPTO \$740.00 International preliminary examination fee (37 CFR 1.482) paid to USPTO but all claims did not satisfy provisions of PCT Article 33(1)-(4) \$710.00 International preliminary examination fee (37 CFR 1.482) paid to USPTO and all claims satisfied provisions of PCT Article 33(1)-(4) \$100.00 ENTER APPROPRIATE BASIC FEE AMOUNT =				CALCULATIONS PTO USE ONLY	
Surcharge of \$130.00 for furnishing the oath or declaration later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(e)).				\$	
CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE	\$	
Total claims	24 - 20 =	4	x \$18.00	\$72.00	
Independent claims	2 - 3 =	0	x \$80.00	\$	
MULTIPLE DEPENDENT CLAIM(S) (if applicable)			+ \$270.00	\$	
TOTAL OF ABOVE CALCULATIONS =				\$962.00	
<input type="checkbox"/> Applicant claims small entity status. See 37 CFR 1.27. The fees indicated above are reduced by ½.				\$	
SUBTOTAL =				\$962.00	
Processing fee of \$130.00 for furnishing the English translation later than <input checked="" type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(f)).				\$	
TOTAL NATIONAL FEE =				\$962.00	
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40.00 per property				\$	
TOTAL FEES ENCLOSED =				\$962.00	
				Amount to be refunded:	\$
				charged:	\$
a. <input type="checkbox"/> A check in the amount of \$ to cover the above fees is enclosed. b. <input checked="" type="checkbox"/> Please charge my Deposit Account No. 12-2475 in the amount of \$962.00 to cover the above fees. A duplicate copy of this sheet is enclosed. c. <input checked="" type="checkbox"/> The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. 12-2475. A duplicate copy of this sheet is enclosed. d. <input type="checkbox"/> Fees are to be charged to a credit card. WARNING: Information on this form may become public. Credit card information should not be included on this form. Provide credit card information and authorization on PTO-2038.					
NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.					
SEND ALL CORRESPONDENCE TO: KRISTIN H. NEUMAN, ESQ. LYON & LYON, LLP SUITE 4700 633 WEST FIFTH STREET LOS ANGELES, CA 90071			SIGNATURE <u>Kristin H. Neuman</u> NAME KRISTIN H. NEUMAN, ESQ. REGISTRATION NUMBER <u>35,530</u>		

10/031135

JC13 Rec'd PCT/PTO 14 JAN 2002

CERTIFICATE OF MAILING
(37 C.F.R. §1.10)

I hereby certify that this paper (along with any referred to as being attached or enclosed) is being deposited with the United States Postal Service on the date shown below with sufficient postage as 'Express Mail Post Office To Addressee' in an envelope addressed to Box PCT, Commissioner for Patents, Washington, D.C. 20231.

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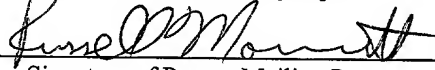
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January 14, 2002

Date of Deposit

Russell Morrisette

Name of Person Mailing Paper



Signature of Person Mailing Paper

2002 JAN 14 10 03 AM

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re: National Stage PCT Application of)
Christoph Rickert et al.)
International Application)
No. PCT/EP00/06423)
Group Art Unit: to be assigned
Examiner: to be assigned
Filed: Concurrently herewith)
Application No.: to be assigned)
35 U.S.C. 371 Date: to be assigned)
For: CURABLE COMPOSITION)

PRELIMINARY AMENDMENT

Box: PCT
Assistant Commissioner for Patents
Washington, D.C. 20231

Dear Sir:

Applicants present this Preliminary Amendment in order to place this National Stage Application in better condition for examination and allowance.

The Commissioner is authorized to charge any fee due, or credit any overcharge, as a result of this Preliminary Amendment to Deposit Account No. 12-2475.

Please amend the National Stage Application as follows:

CERTIFICATE OF MAILING
(37 C.F.R. §1.10)

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Russell Morrisette

Name of Person Mailing Paper

Signature of Person Mailing Paper

IN THE CLAIMS:

Please cancel claims 1-17, and add the following new claims 18-41.

18. (new) A curable composition comprising:

(A) from 80 to 97.9 percent by weight, based on the total amount of components (A), (B) and (C), of a polyester component having an acid number in total of from 10 to 40 mg of KOH per gram of the component and consisting of one or more polyesters containing free carboxyl groups;

(B) from 0.2 to 2 percent by weight, based on the total amount of components (A), (B) and (C), of a component consisting of one or more polymers or copolymers that each have an epoxy value of at least 3 equivalents per kilogram and a molecular weight (number average M_n) of from 800 to 20,000 and that contain identical or different structural repeating units of formula (I):



wherein R¹ and R² are each independently of the other a hydrogen atom or a methyl group; and

(C) a component consisting of one or more monomeric polyglycidyl compounds having an epoxy value of at least 3.5 equivalents per kilogram and a maximum molecular weight of 600, the amount of component (C) in percent by weight, based on the total amount of components (A), (B) and (C), corresponding to the difference between 100 and the sum of the percentage amounts of components (A) and (B) based on the total amount of components (A), (B) and (C); and

(D) an accelerator for the reaction of epoxy groups with carboxyl groups.

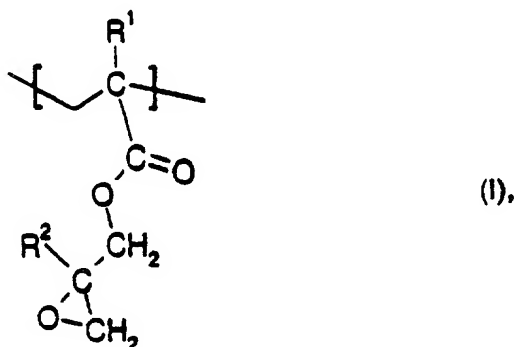
19. (new) A composition according to claim 18 that contains from 85 to 96 percent by weight of component (A) based on the total amount of components (A), (B) and (C).
20. (new) A composition according to claim 19 that contains from 88 to 95 percent by weight of component (A) based on the total amount of components (A), (B) and (C).
21. (new) A composition according to claim 18 in which the polyester component (A) has an acid number in total of from 22 to 38 mg of KOH per gram of the component.
22. (new) A composition according to claim 18 in which the polyester(s) that constitute the polyester component (A) have a molecular weight of from 4,000 to 15,000 (weight average Mw from GPC measurement using polystyrene calibration) and a glass transition temperature (Tg) of from 35 to 120°C.
23. (new) A composition according to claim 22 in which the polyester(s) that constitute the polyester component (A) have a Mw of from 6,500 to 11,000 and a Tg of from 50 to 90°C.
24. (new) A composition according to claim 18 in which the polyester(s) that constitute the polyester component (A) are based on neopentyl glycol and/or trimethylolpropane as the main alcoholic monomer constituent(s) and on adipic acid and/or terephthalic acid and/or isophthalic acid and/or trimellitic acid as the main acidic monomer component(s).
25. (new) A composition according to claim 18 that contains from 0.7 to 2 percent by weight of component (B) based on the total amount of components (A), (B) and (C).
26. (new) A composition according to claim 25 that contains from 0.7 to 1.8 percent by weight of component (B) based on the total amount of components (A), (B) and (C).
27. (new) A composition according to claim 18 in which component (B) constitutes from 5 to 50 percent of the total weight of components (B) and (C).

28. (new) A composition according to claim 27 in which component (B) constitutes from 7.5 to 25 percent of the total weight of components (B) and (C).

29. (new) A composition according to claim 18 in which component (B) consists of one or more polymers or copolymers each of which has an epoxy value of at least 5 equivalents per kilogram.

30. (new) A composition according to claim 18 in which the polymers and copolymers that constitute component (B) have a molecular weight (number average Mn) of from 1,000 to 10,000.

31. (new) A composition according to claim 18 in which component (B) is constituted by polymers that consist substantially of identical or different structural repeating units of formula (I):



wherein the radicals R^1 and R^2 are each independently of the other a hydrogen atom or a methyl group.

32. (new) A composition according to claim 18 wherein R^2 in formula (I) is a hydrogen atom.

33. (new) A composition according to claim 18 that contains from 0.1 to 38 percent by weight of component (C) based on the total amount of components (A), (B) and (C).

34. (new) A composition according to claim 33 that contains from 5 to 15 percent by weight of component (C) based on the total amount of components (A), (B) and (C).
35. (new) A composition according to claim 18 in which component (C) consists of one or more polyglycidyl compounds having an epoxy value of at least 5 equivalents per kilogram.
36. (new) A composition according to claim 18 in which component (C) consists of one or more monomeric polyglycidyl compounds selected from trimellitic acid triglycidyl ester, triglycidyl isocyanurate, hexahydroterephthalic acid diglycidyl ester, hexahydroisophthalic acid diglycidyl ester, hexahydrotrimellitic acid triglycidyl ester, terephthalic acid diglycidyl ester, isophthalic acid diglycidyl ester and cyclohexanedimethanol diglycidyl ether.
37. (new) A composition according to claim 36 in which component (C) consists of transhexahydroterephthalic acid diglycidyl ester.
38. (new) A composition according to claim 18 in which the epoxy value of components (B) and (C) together is at least 3.5 equivalents per kilogram.
39. (new) A composition according to claim 38 in which the epoxy value of components (B) and (C) together is at least 5 equivalents per kilogram.
40. (new) A composition according to claim 18 in which the molar ratio of glycidyl groups to carboxyl groups in the composition is from 1.3:1 to 1:1.
41. (new) A powder coating composition based on a curable composition according to claim 18.

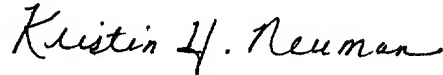
REMARKS

Claims 1-17 are present in the above-referenced International Application, upon which this National Stage Application is based. New claims 18-41 contain the same subject matter as

original claims 1-17, but have been rewritten to eliminate the multiple dependencies. Accordingly, no new matter has been added.

In view of the foregoing amendments, the new claims are now in better condition for examination and allowance.

Respectfully submitted



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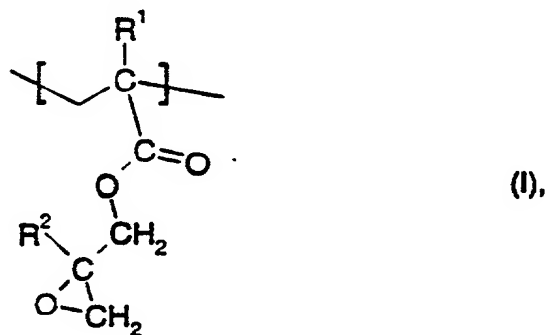
Curable composition

The invention relates to a curable composition based on acidic polyesters and on certain polyglycidyl compounds and to a powder coating composition based thereon.

JP-A-Sho 57-108166 discloses a curable composition and the use thereof as a powder coating composition, the composition comprising

- (A) from 60 to 96 parts by weight of a polyester having an acid number of from 20 to 200 mg of KOH per gram, a softening point of from 80 to 150 °C and a molecular weight (number average) of from 1000 to 10 000,
- (B) from 3 to 40 parts by weight of an acrylic polymer containing glycidyl groups and having a molecular weight (number average) of from 300 to 5000,
- (C) from 1 to 20 parts by weight of a further epoxy resin, which may be, *inter alia*, diglycidyl terephthalate or triglycidyl isocyanurate, and
- (D) an accelerator for the reaction of epoxy groups with carboxyl groups.

There are used as component (B) polymers or copolymers that contain identical or different structural repeating units of formula (I):



wherein the radicals R¹ and R² are each independently of the other a hydrogen atom or a methyl group.

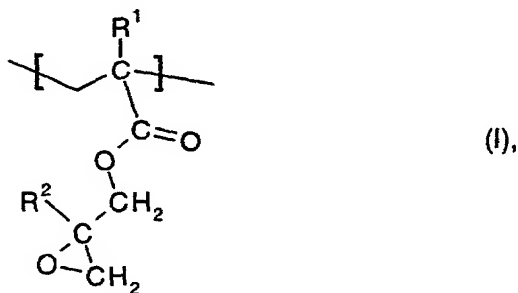
As can be seen from the above quantitative data, the said compositions contain at least approximately 2.5 % by weight of component (B), based on the total amount of components (A), (B) and (C). The flow behaviour of the compositions is generally still not satisfactory, however, and accordingly when such compositions are used in the production of powder

resin coatings there is, in particular, a risk that the known orange-peel effect will occur, that is to say, that a coating having a very uneven surface will be formed.

The present invention is based on the surprising discovery that the flow behaviour of the above-mentioned curable compositions is substantially improved but, nevertheless, cured material having excellent physical properties, especially excellent mechanical properties, is obtainable by using as component (B) of the compositions a maximum of 2 percent by weight, based on the total amount of components (A), (B) and (C), of one or more polymers or copolymers that each have an epoxy value of at least 3 equivalents per kilogram and a molecular weight (number average M_n) of from 800 to 20 000 and that contain identical or different structural repeating units of the formula (I) already mentioned above.

The present invention accordingly relates to a curable composition comprising:

- (A) from 80 to 97.9 percent by weight, based on the total amount of components (A), (B) and (C), of a polyester component having an acid number in total of from 10 to 40 mg of KOH per gram of the component and consisting of one or more polyesters containing free carboxyl groups;
- (B) from 0.2 to 2 percent by weight, based on the total amount of components (A), (B) and (C), of a component consisting of one or more polymers or copolymers that each have an epoxy value of at least 3 equivalents per kilogram and a molecular weight (number average M_n) of from 800 to 20 000 and that contain identical or different structural repeating units of formula (I):



wherein R^1 and R^2 are each independently of the other a hydrogen atom or a methyl group; and

- (C) a component consisting of one or more monomeric polyglycidyl compounds having an epoxy value of at least 3.5 equivalents per kilogram and a maximum molecular weight of 600, the amount of component (C) in percent by weight, based on the total amount

of components (A), (B) and (C), corresponding to the difference between 100 and the sum of the percentage amounts of components (A) and (B) based on the total amount of components (A), (B) and (C); and

(D) an accelerator for the reaction of epoxy groups with carboxyl groups, wherein, furthermore, component (B) constitutes from 5 to 95 percent of the total weight of components (B) and (C) and the molar ratio of glycidyl groups to carboxyl groups in the composition is from 1.3:1 to 1:1.3.

Curable compositions according to the present invention are distinguished, *inter alia*, by a very good flow behaviour and yield a cured material that has a high crosslinking density and excellent mechanical properties.

The curable compositions according to the invention are suitable, for example, as adhesives, casting resins or compression-moulding compositions, it being possible for further components that are conventional in the field of use in question to be admixed therewith, as desired.

Special preference is given to powder coating compositions that contain as curable component system the compositions according to the invention. The invention accordingly relates also to powder coating compositions based on the curable compositions according to the invention.

The polyester component (A) constitutes preferably from 85 to 96 percent by weight, especially from 88 to 95 percent by weight, of the composition according to the invention based on the total amount of components (A), (B) and (C).

The acid number of from 10 to 40 mg of KOH per gram applies to the polyester component as a whole, and therefore corresponds, when several different polyesters are used, to the average acid number of the totality of those polyesters. Preference is given to compositions according to the invention in which the polyester component (A) has an acid number in total of from 20 to 40 mg, preferably from 22 to 38 mg, of KOH per kilogram of the component.

The polyesters of component (A) are advantageously solid at room temperature (from 15 to 35°C) and have, for example, a molecular weight (number average M_n) of from 1000 to

10 000. The ratio of Mw (weight average of the molecular weight) to Mn of those polyesters is generally from 2 to 10. There are especially suitable for the present invention, for example, free-carboxyl-group-containing polyesters having a molecular weight (weight average Mw from GPC measurement using polystyrene calibration) of from 4000 to 15 000, especially from 6500 to 11 000, and a glass transition temperature (Tg) of from 35 to 120°C, preferably from 50 to 90°C.

Polyesters such as those mentioned are described, for example, in US-A-3 397 254 and EP-A-0 600 546. Polyesters suitable for the present invention are condensation products of difunctional, trifunctional and/or polyfunctional alcohols (polyols) with dicarboxylic acids and, optionally, trifunctional and/or polyfunctional carboxylic acids, or with corresponding carboxylic acid anhydrides. The polyols used include, for example, ethylene glycol, diethylene glycol, the propylene glycols, butylene glycol, 1,3-butanediol, 1,4-butanediol, neopentanediol, isopentyl glycol, 1,6-hexanediol, glycerol, hexanetriol, trimethylolethane, trimethylolpropane, erythritol, pentaerythritol, cyclohexanediol and 1,4-dimethylolcyclohexane. Suitable dicarboxylic acids include, for example, isophthalic acid, terephthalic acid, phthalic acid, methyl-substituted derivatives of the said acids, tetrahydrophthalic acid, methyl-tetrahydrophthalic acids, for example 4-methyltetrahydrophthalic acid, cyclohexane-dicarboxylic acids, succinic acid, glutaric acid, adipic acid, pimelic acid, suberic acid, azelaic acid, sebacic acid, dodecanedicarboxylic acid, fumaric acid, maleic acid and 4,4'-diphenyldicarboxylic acid etc.. Suitable tricarboxylic acids include, for example, aliphatic tricarboxylic acids, such as 1,2,3-propanetricarboxylic acid, aromatic tricarboxylic acids, such as trimesic acid, trimellitic acid and hemimellitic acid, and cycloaliphatic tricarboxylic acids, such as 6-methylcyclohex-4-ene-1,2,3-tricarboxylic acid. Suitable tetracarboxylic acids include, for example, pyromellitic acid and benzophenone-3,3',4,4'-tetracarboxylic acid. Commercially available polyesters especially are very commonly based on neopentyl glycol and/or trimethylolpropane as the main alcoholic monomer constituent(s) and on adipic acid and/or terephthalic acid and/or isophthalic acid and/or trimellitic acid as the main acidic monomer component(s).

Component (B) constitutes preferably from 0.7 to 2 percent by weight, especially from 0.7 to 1.8 percent by weight, of the composition according to the invention based on the total weight of components (A), (B) and (C). It is furthermore preferred for component (B) to

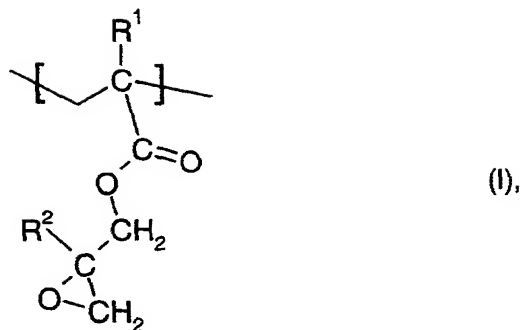
constitute from 5 to 50 percent, especially from 7.5 to 25 percent, of the total weight of the polyglycidyl components (B) and (C).

The polymers and copolymers constituting component (B) of the compositions according to the invention preferably have a molecular weight (number average M_n) of from 1000 to 10 000. The polymers and copolymers can be prepared, for example, by a conventional free-radical polymerisation of a monomer selected from glycidyl acrylate, glycidyl methacrylate, β -methylglycidyl acrylate and β -methylglycidyl methacrylate, optionally in the presence of a suitable amount of free-radically polymerisable comonomer(s). The polymerisation can be carried out, for example, by dissolving the monomers in a suitable organic solvent, such as toluene, 1-methoxy-2-propanol, 1-methoxy-2-propyl acetate or methyl isobutyl ketone, or in a mixture of the said solvents, and heating the resulting solution in the presence of a suitable initiator, such as, for example, α,α' -azoisobutyronitrile or dicumyl peroxide, optionally in the presence of a chain terminator, such as, for example, allyl glycidyl ether. Suitable comonomers include, for example, (meth)acrylic acid esters, such as ethyl acrylate, butyl acrylate, 2-ethyl hexylacrylate, and also, especially, C_1 - C_4 alkyl methacrylates, such as methyl methacrylate, ethyl methacrylate and butyl methacrylate, and furthermore, for example, acrylo- and methacrylo-nitriles, (meth)acrylamides and also vinyl compounds, for example aromatic vinyl compounds, especially styrene and styrene derivatives.

It is also possible, however, to glycidylise a poly(meth)acrylic acid polymer or a poly(meth)-acrylic acid copolymer according to one of the conventional methods, for example by reacting the polymer or copolymer with an excess of epichlorohydrin in the presence of a suitable catalyst, after which the resulting polyhalohydrin esters are dehydrochlorinated using a hydrogen-chloride-binding agent.

Component (B) consists preferably of polymers or copolymers each having an epoxy value of at least 5 equivalents per kilogram.

It is more especially preferred for component (B) to be constituted by polymers that consist substantially of identical or different structural repeating units of formula (I):



wherein R^1 and R^2 are each independently of the other a hydrogen atom or a methyl group, especially when R^2 in formula (I) is a hydrogen atom. Finally, the polyglycidyl component (C) constitutes preferably from 0.1 to 38 percent by weight, especially from 5 to 15 percent by weight, of the composition according to the invention based on the total amount of components (A), (B) and (C).

Component (C) consists of one or more polyglycidyl or poly(β -methyl)glycidyl compounds each having an epoxy value of at least 3.5 equivalents per kilogram. Those compounds, in contrast to the polyglycidyl compounds constituting component (B) of the compositions according to the invention, are monomeric compounds having a maximum molecular weight of 600 (molecular weight being understood in this case to mean the sum, calculated theoretically for each of the compounds, of the weights of the atoms of the molecule), for example corresponding diglycidyl or triglycidyl compounds that are preferably solid at room temperature, or corresponding polyglycidyl compounds having an even larger number of glycidyl groups per molecule.

Preferably, component (C) of the compositions consists of one or more glycidyl compounds having an epoxy value of at least 4 equivalents, especially at least 5 equivalents, per kilogram.

Examples of glycidyl compounds that are suitable as component (C) of the compositions according to the invention include, *inter alia*, trimellitic acid triglycidyl ester, triglycidyl isocyanurate, hexahydroterephthalic acid diglycidyl ester, especially *trans*-hexahydroterephthalic acid diglycidyl ester, hexahydroisophthalic acid diglycidyl ester, hexahydrotrimellitic acid triglycidyl ester, terephthalic acid diglycidyl ester, isophthalic acid diglycidyl ester and cyclohexanedimethanol diglycidyl ether.

For the composition according to the invention it is furthermore preferred for the epoxy value of components (B) and (C) together likewise to be at least 3.5 equivalents, preferably at least 5 equivalents, per kilogram of the mixture of the two components.

The compositions according to the invention preferably have a slight molar excess of epoxy groups. The molar ratio of glycidyl groups to carboxyl groups in the compositions is thus preferably from 1.3:1 to 1:1, for example approximately from 1.2:1 to 1.1:1.

In order that the curing reaction can proceed sufficiently rapidly, the compositions according to the invention also contain, as component (D), a catalyst for the reaction of epoxy groups with carboxyl groups. Such a catalyst is frequently an organic amine or a derivative of an amine, especially a tertiary amine or a nitrogen-containing heterocyclic compound. Preferred catalysts for the reaction of epoxy groups with carboxyl groups are phenylimidazole, N-benzyltrimethylamine and 1,8-diazabicyclo[5.4.0]-7-undecene, optionally on a silicate support or triphenylphosphine, Actiron® NXJ-60 (2-propylimidazole), Actiron® NXJ-60 P (60 % by weight of 2-propylimidazole on 40 % by weight of solid support), Beschleuniger® DT 3126 ($[\text{C}_{16}\text{H}_{33}\text{N}(\text{CH}_3)_3]^+\text{Br}^-$). The catalyst or a catalyst mixture is preferably added in such an amount that the gel time of the mixture at 180°C (determined according to DIN 55990) is approximately from 70 to 400 seconds, preferably from 90 to 300 seconds. Generally, approximately from 0.1 to 10 percent by weight, especially from 0.5 to 5 percent by weight, of catalyst will be required for that purpose. Of course some commercially available polyesters that are used as a constituent of component (A) of the compositions according to the invention will already contain a certain amount of one of the above-mentioned catalysts or of a comparable catalyst, and that amount should be taken into account in the above percentage by weight figure for catalyst; the mentioned preferred gel times can be used to provide an indication of how much catalyst still needs to be added.

In addition to the components already mentioned, the compositions according to the invention may, of course, contain further components, which may vary according to the field of application of the compositions and which will be known to the person skilled in the art in question.

Powder coating compositions based on the compositions according to the invention may, for example, also contain additives customary in the surface-coating industry in the amounts conventionally used therefor, such as, *inter alia*, light stabilizers, curing accelerators, dyes, pigments, for example titanium dioxide pigment, degassing agents, for example benzoin, and also additional flow agents. Suitable flow agents include, for example, polyvinyl acetals, such as polyvinyl butyral, polyethylene glycol, polyvinylpyrrolidone, glycerol and acrylic mixed polymers, such as, for example, those available under the names Modaflow® and Acrylron®.

The powder coating compositions according to the invention can be prepared simply by mixing the constituents together, for example in a ball mill. Another, more preferred possibility comprises melting together, blending and homogenising the constituents, preferably using an extrusion machine, such as a Buss co-kneader, and cooling and comminuting the resulting mass. In that procedure, the fact that either immediately after extrusion, or at least after they have been left to stand for a few hours, for example from 24 to 48 hours, the powder coating compositions according to the invention become so hard and brittle that they can readily be ground, has proved especially advantageous. The powder coating mixtures preferably have a particle size in the range from 0.015 to 500 µm, especially from 10 to 75 µm. In some cases it may also be advantageous first of all to prepare a masterbatch from portions of the binder, the epoxy resins and, optionally, further components, the masterbatch then being mixed and homogenised in a second step with the remainder of the binder and the remaining constituents to yield the finished powder coating composition.

After application to the article to be coated, the powder coating compositions are cured at a temperature of at least approximately 100°C, preferably at from 150 to 220°C. Curing generally takes approximately from 5 to 60 minutes. All materials that are stable at the temperatures required for the curing, especially ceramics and metals, are suitable for coating. The substrate may already have one or more base surface-coatings that are compatible with the powder coating composition.

The powder coating compositions exhibit a good flow behaviour combined with good mechanical properties, good weather resistance and good resistance to chemicals.

acetone/water. The solid polymer is filtered off and dried *in vacuo*. 1200.00 g of colourless polymer in powder form, having the following analytical data, remain:

T_g , measured by DSC : 31°C

GPC (polystyrene calibration): $M_w = 7147$; $M_n = 2986$

Epoxy value, titrimetric: 6.44 equivalents/kg

Example 2: Preparation of a further GMA homopolymer

The following are polymerised in accordance with the procedure indicated in Example 1:

Monomer mixture: 144.50 g of glycidyl methacrylate + 25.50 g of allyl glycidyl ether

Initial reactor charge: 17.00 g of monomer mixture + 102.00 g of MPA

Feed: 153.00 g of monomer mixture + 11.30 g of MPA + 8.50 g of AIBN

158.30 g of colourless polymer in powder form, having the following analytical data, are obtained:

T_g : 42°C

GPC (polystyrene calibration): $M_w = 6099$; $M_n = 1304$

Epoxy value, titrimetric: 6.39 equivalents/kg

Example 3: Preparation of a GMA copolymer (Rt 2549/1)

The following are polymerised in accordance with the procedure indicated in Example 1:

Monomer mixture: 25.50 g of butyl methacrylate + 127.50 g of glycidyl methacrylate + 17.00 g of allyl glycidyl ether

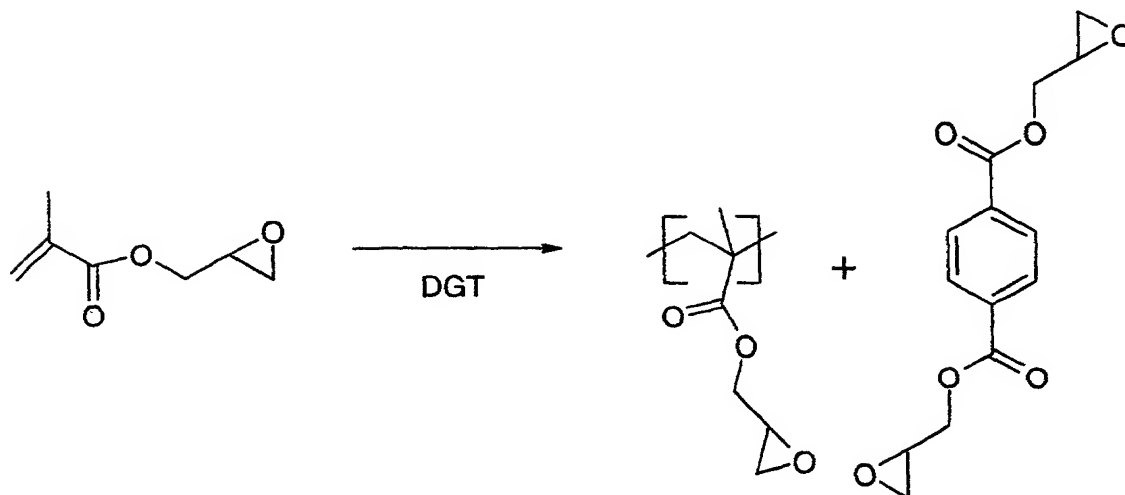
Initial reactor charge: 17.00 g of monomer mixture and 102.00 g of MPA

Feed: 153.00 g of monomer mixture + 11.30 g of MPA + 8.50 g of AIBN

The isolation of the solid polymer is slightly modified: the polymer solution is concentrated to dryness using a rotary evaporator, the residue is dissolved in acetone and then precipitated from ice-water. 157.00 g of colourless polymer in powder form, having the following analytical data, are obtained:

GPC (polystyrene calibration): $M_w = 7182$; $M_n = 1926$

Epoxy value, titrimetric: 5.47 equivalents/kg

Example 4: Free-radical polymerisation of GMA in diglycidyl terephthalate (DGT)

200 g of diglycidyl terephthalate (DGT) are introduced into a 1 litre flat ground flask. 40 g of glycidyl methacrylate (GMA) and 4 g of 1,1-bis(tert-butylperoxy)-3,3,5-trimethylcyclohexane (50% solution = Trigonox 29, C50 from AKZO) are introduced into a 250 ml taper-necked flask. The two flasks are connected to each other. The flat ground flask is heated by means of an oil bath until the DGT has melted. When all of the DGT has melted, the 1 litre flat ground flask, together with the 250 ml supply flask connected to it, is evacuated and filled with nitrogen a total of five times. The mixture of GMA and 1,1-bis(tert-butylperoxy)-3,3,5-trimethylcyclohexane is then fed from the 250 ml supply flask into the molten DGT within a period of 20 minutes, with stirring, and the batch is left to react for 3.5 hours with further stirring. On subsequent cooling, the liquid mass solidifies.

Yield: 236 g (98%)

GPC (polystyrene calibration): $M_w = 15\,700$; $M_n = 8921$

Epoxy value, titrimetric: 6.86 equivalents/kg

Example 5:

Using an extruder (laboratory extruder from PRISM, The Old Stables, England), the powder coating compositions indicated in the following Tables 1a and 1b are homogenised at a temperature of from 60 to 80°C. The total amount of powder coating composition in each case is approximately from 100 to 200 grams. The cooled extrudates are ground to give the finished powder coating composition having a particle size of approximately 40 μm .

Table 1a: Powder coating composition formulations A to C according to the invention and comparison coating compositions E to G

Formulation/ amounts [% by wt.]	Invention			Comparison		
	A	B	C	E	F	G
Uralac P 3485 ¹⁾	58.98	59.96	90.77	59.08	59.81	59.95
DGT	4.69	3.97	6.01	4.89	---	---
GMA polymer of Ex. 1	---	1.03	---	---	5.16	---
GMA polymer of Ex. 2	---	---	1.52	---	---	5.02
GMA polymer (6.7 eq/kg)	0.80					
DT 3126-1 ²⁾	0.50	---	---	1.00	---	---
Benzoin	0.20	0.20	0.20	0.20	0.20	0.20
Acrylon ³⁾	1.50	1.50	1.50	1.50	1.50	1.50
TiO ₂ pigment	33.33	33.33	---	33.33	33.33	33.33

¹⁾ Polyester based on terephthalic acid, isophthalic acid and neopentyl glycol having an acid number of 28 and a glass transition temperature T_g of 71°C;

²⁾ Alkylammonium salt in polyester;

³⁾ Acrylic mixed polymer as flow agent.

Table 1b: Powder coating composition formulations H to K and P according to the invention

Formulation [% by wt.]	Invention				
	H	I	J	K	P
Polyester ⁴⁾	<i>Uralac P-3485</i> 58.71	<i>Crylcoat 801</i> 57.61	<i>Crylcoat D-6210</i> 59.66	<i>Alftalat 9952</i> 58.90	<i>Uralac P-3485</i> 59.09
DGT/GMA mixed polymer	5.76 ⁵⁾	6.86 ⁵⁾	4.81 ⁵⁾	5.57 ⁵⁾	5.38 ⁶⁾
Benzoin	0.20	0.20	0.20	0.20	0.50
Acrylon	1.50	1.50	1.50	1.50	0.20
XB 3126	0.50	0.50	0.50	0.50	1.50
TiO ₂ pigment	33.33	33.33	33.33	33.33	33.33

⁴⁾ Polyester based on terephthalic acid, isophthalic acid and neopentyl glycol having an acid number and a glass transition temperature according to the following Table:

Polyester	<i>Crylcoat 801</i>	<i>Crylcoat D-6210</i>	<i>Alftalat 9952</i>
Acid number	34	23	27
T _g (°C)	73	72	64

*First DSC run.

⁵⁾ Mixture of approximately 86 % by weight DGT and 14 % by weight GMA homopolymer having an epoxy value of 6.37 equivalents per kilogram of the mixture.

⁶⁾ Mixture of DGT and GMA homopolymer according to Example 4.

Using an electrostatic spray pistol, the powder coating compositions are applied to Q panels as substrate. The coated panels are then placed in an oven in order to melt and fully cure the powder coating composition. The gel time, the curing temperature and the curing time,

and also the thickness of the resulting powder resin coating, are indicated in the following Tables 2a and 2b together with properties of the resulting coatings that are important from the standpoint of surface-coating technology.

Table 2a: Properties, important from the standpoint of surface-coating technology, of the coatings obtained using compositions A, B, C and E, F, G

	Invention			Comparison		
	A	B	C	E	F	G
Gel time ⁸⁾ at 180°C [s]	220	170	90	205	25	20
Curing conditions	15 min./180°C	15 min./180°C	15 min./180°C	15 min./180°C	15 min./180°C	15 min./180°C
Layer thickness [µm]	60	53	59	60	59	60
60° gloss	94	96	124	87	18	21
20° gloss	85	77	117	95	60	65
Yellowness value Yi ⁷⁾	0.2	-0.8	---	-1.2	-1.1	-0.8
Flow ⁸⁾ [rating]	10	10	12	12	orange-peel	orange-peel
Impact deformation, reverse ⁹⁾ [kg cm]	160	160	160	160	<5	<5
Impact deformation, front [kg cm]	160	160	140	160	80	80
Acetone test, ¹⁰⁾ 1 min. [rating]	3	3	3	5 not cured	3	3
Appearance	satisfactory	satisfactory	satisfactory	satisfactory	mat	mat

⁸⁾ Empirical scale from 0 (very good) to 18 (orange-peel)

⁹⁾ The impact deformation is determined by dropping onto the coated face a punch weighing 2 kg, having a 20 mm diameter ball on its underside, with the underside leading, from a specific height from behind (reverse side) or from the front. The value indicated is the product of the weight of the punch in kg and the test height in cm at which there is still no detectable damage to the coating.

¹⁰⁾ According to DIN 53320. The specimen is kept in acetone for 1 minute. The result is evaluated in accordance with the following scale of five ratings: 0 = unchanged; 1 = resistant, cannot be scratched with a finger nail; 2 = difficult to scratch, may stain cottonwool pad; 3 = softened, easily scratchable; 4 = beginning to separate or dissolve; 5 = complete dissolution.

Table 2b: Surface-coating technology properties of the coatings using compositions

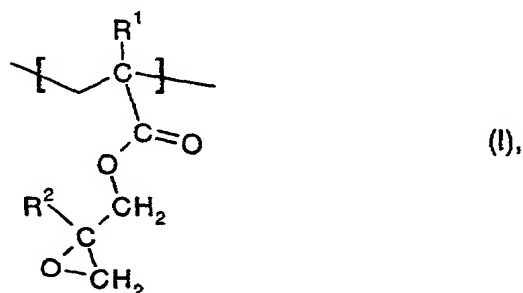
H to K, P

	Invention				
	H	I	J	K	P
Gel time at 180°C [s]	225	275	180	170	260
Curing conditions	15 min. / 200°C	15 min. / 200°C	15 min. / 200°C	15 min. / 200°C	15 min. / 180°C
Layer thickness [μm]	60	55	50	47	46
60° gloss	89	95	94	96	95
20° gloss	76	88	84	83	85
Yellowness value Y _i	-0.4	0.0	-1.0	-1.8	-1.5
Flow [rating]	8	8	10	10	10
Impact deformation, reverse [kg cm]	>160	120	>160	>160	>160
Impact deformation, direct [kg cm]	>160	>160	>160	>160	>160
Acetone test, 1 min [rating]	3	3	3	3	3
Appearance	satisfactory	satisfactory	satisfactory	satisfactory	satisfactory

What is claimed is:

1. A curable composition comprising:

- (A) from 80 to 97.9 percent by weight, based on the total amount of components (A), (B) and (C), of a polyester component having an acid number in total of from 10 to 40 mg of KOH per gram of the component and consisting of one or more polyesters containing free carboxyl groups;
- (B) from 0.2 to 2 percent by weight, based on the total amount of components (A), (B) and (C), of a component consisting of one or more polymers or copolymers that each have an epoxy value of at least 3 equivalents per kilogram and a molecular weight (number average Mn) of from 800 to 20 000 and that contain identical or different structural repeating units of formula (I):



wherein R¹ and R² are each independently of the other a hydrogen atom or a methyl group; and

- (C) a component consisting of one or more monomeric polyglycidyl compounds having an epoxy value of at least 3.5 equivalents per kilogram and a maximum molecular weight of 600, the amount of component (C) in percent by weight, based on the total amount of components (A), (B) and (C), corresponding to the difference between 100 and the sum of the percentage amounts of components (A) and (B) based on the total amount of components (A), (B) and (C); and
- (D) an accelerator for the reaction of epoxy groups with carboxyl groups

2. A composition according to claim 1 that contains from 85 to 96 percent by weight, especially from 88 to 95 percent by weight, of component (A) based on the total amount of components (A), (B) and (C).

3. A composition according to either claim 1 or claim 2 in which the polyester component (A) has an acid number in total of from 22 to 38 mg of KOH per gram of the component.

4. A composition according to any one of claims 1 to 3 in which the polyesters that constitute the polyester component (A) have a molecular weight of from 4000 to 15 000, especially from 6500 to 11 000 (weight average Mw from GPC measurement using polystyrene calibration) and a glass transition temperature (Tg) of from 35 to 120°C, preferably from 50 to 90°C.

5. A composition according to any one of claims 1 to 4 in which the polyesters that constitute the polyester component (A) are based on neopentyl glycol and/or trimethylolpropane as the main alcoholic monomer constituent(s) and on adipic acid and/or terephthalic acid and/or isophthalic acid and/or trimellitic acid as the main acidic monomer component(s).

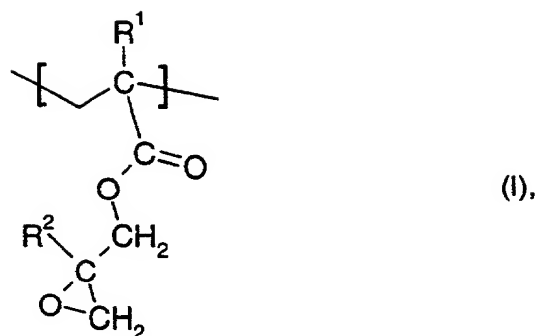
6. A composition according to any one of claims 1 to 5 that contains from 0.7 to 2 percent by weight, especially from 0.7 to 1.8 percent by weight, of component (B) based on the total amount of components (A), (B) and (C).

7. A composition according to any one of claims 1 to 6 in which component (B) constitutes from 5 to 50 percent, especially from 7.5 to 25 percent, of the total weight of components (B) and (C).

8. A composition according to any one of claims 1 to 7 in which component (B) consists of one or more polymers or copolymers each of which has an epoxy value of at least 5 equivalents per kilogram.

9. A composition according to any one of claims 1 to 8 in which the polymers and copolymers that constitute component (B) have a molecular weight (number average Mn) of from 1000 to 10 000.

10. A composition according to any one of claims 1 to 9 in which component (B) is constituted by polymers that consist substantially of identical or different structural repeating units of formula (I):



wherein the radicals R¹ and R² are each independently of the other a hydrogen atom or a methyl group.

11. A composition according to any one of claims 1 to 10 wherein R² in formula (I) is a hydrogen atom.
12. A composition according to any one of claims 1 to 11 that contains from 0.1 to 38 percent by weight, preferably from 5 to 15 percent by weight, of component (C) based on the total amount of components (A), (B) and (C).
13. A composition according to any one of claims 1 to 12 in which component (C) consists of one or more polyglycidyl compounds having an epoxy value of at least 5 equivalents per kilogram.
14. A composition according to any one of claims 1 to 13 in which component (C) consists of one or more monomeric polyglycidyl compounds selected from trimellitic acid triglycidyl ester, triglycidyl isocyanurate, hexahydroterephthalic acid diglycidyl ester, especially *trans*-hexahydroterephthalic acid diglycidyl ester, hexahydroisophthalic acid diglycidyl ester, hexahydrotrimellitic acid triglycidyl ester, terephthalic acid diglycidyl ester, isophthalic acid diglycidyl ester and cyclohexanedimethanol diglycidyl ether.
15. A composition according to any one of claims 1 to 14 in which the epoxy value of components (B) and (C) together is at least 3.5 equivalents, preferably at least 5 equivalents, per kilogram.

16. A composition according to any one of claims 1 to 15 in which the molar ratio of glycidyl groups to carboxyl groups in the composition is from 1.3:1 to 1:1.

17. A powder coating composition based on a curable composition according to anyone of claims 1 to 16.

**UTILITY DECLARATION
AND POWER OF ATTORNEY
Utility Application**

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name.

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled

Curable composition

the specification of which

- ☐ is attached hereto OR
☐ was filed on _____ as United States Application Serial No. ____ OR
☒ was filed on 06 July 2000 as PCT International Application No. PCT/EP 00/06423
☐ and was amended on ____ (if applicable).

I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment(s) referred to above.

I acknowledge the duty to disclose information which is material to the patentability of this application in accordance with Title 37, Code of Federal Regulations, § 1.56.

I hereby claim foreign priority benefits under Title 35, United States Code, § 119(a)-(d) or § 365(b) of any foreign application(s) for patent or inventor's certificate, or § 365(a) of any PCT international application which designated at least one country other than the United States of America, listed below and have also identified below, by checking the box, any foreign application for patent or inventor's certificate, or of any PCT international application having a filing date before that of the application on which priority is claimed.

Prior Foreign Application Number(s)	Country	Date of Filing (day/month/year)	Priority Claimed	
			Yes	No
1304/99	Switzerland	14/07/1999	<input checked="" type="checkbox"/>	<input type="checkbox"/>

I hereby claim the benefit under Title 35, United States Code § 119(e) of any United States provisional application(s) listed below.

Application Number(s)	Filing Date (day/month/year)

I hereby claim the benefit under Title 35, United States Code, § 120 of any United States application(s), or § 365(c) of any PCT international application designating the United States of America, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT international application in the manner provided by the first paragraph of Title 35, United States Code, § 112, I acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations, § 1.56 which became available between the filing date of the prior application and the national or PCT international filing date of this application.

U.S. Parent Application Number	PCT Parent Number	Parent Filing Date (day/month/year)	Status-Patented, Pending or Abandoned

POWER OF ATTORNEY: As a named inventor, I hereby appoint as my attorneys and/or agents, with full power of substitution and revocation, to prosecute this application and transact all business in the United States Patent and Trademark Office, and in countries other than the United States, and to do all things necessary or appropriate therefor before any competent International Authorities in connection with any international patent application(s) corresponding to the above-identified invention application, all of the registered practitioners identified by Customer-Number 22249:



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I further declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements are made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Title 18, United States Code, § 1001 and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

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